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Optical Phase Control of Ultrashort Femtosecond Pulses

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ABSTRACT

When optical pulses achieve durations as short as a few optical cycles, their propagation and nonlinear interaction properties will begin to depend on the phase of the optical waveform relative to its envelope. We have recently demonstrated the feasibility of controlling the relative optical phase of successive pulses from a laser and propose here to study the effect of that control on the pulse formation process itself as well as on other nonlinear optical processes. Phase stabilized Ti:sapphire and Cr:YAG lasers will be developed for experiments in two different wavelength regimes.

PROPOSAL

Ultrashort pulses of less than 10fs duration hold considerable promise for extending the investigation of ultrafast optically active processes into an interesting new regime. The shortest pulses obtained thus far, directly from a laser oscillator are 6 fs long [1]. Pulses as short as 4.5 fs have been produced by subsequent pulse compression, by the Wiersma group in the Netherlands [2] and by a collaboration between groups in Vienna, Milan and Budapest [3]. At the 850nm wavelength of the Ti:sapphire laser, 3 fs corresponds to about one wavelength. So, these pulses are in the regime of two cycles or less. Their optical phase should begin to be important to their formation and to their subsequent nonlinear interactions.

A freely propagating pulse consisting of many optical cycles may to good approximation be described by an amplitude envelope function that does not depend upon the phase of the optical wave it modulates. When the pulse propagates in a medium in which the group velocity differs from the phase velocity, the optical phase slides relative to the envelope which is relatively unaffected by this sliding. If the pulse only contains a few optical cycles, this is no longer true. In order to gain some appreciation for this sensitivity of ultrashort pulses to the optical phase consider a simple comparison. If we assume that there can be no zero frequency component in a freely propagating wave, then $A(t)\sin\omega t$ always has an acceptable spectrum, but $A(t)\cos\omega t$ may not. Thus, it is apparent that the phase of the pulse cannot change arbitrarily by itself without changing the envelope. Otherwise the spectrum would change, especially at low frequencies. This implies that as $A(t)\sin\omega t$ propagates in a dispersive medium it cannot simply accumulate a frequency independent optical phase shift. Instead it develops some chirp; the zero crossings of the field as a function of time become nonuniformly spaced. One particular

phase evolution that works is one that varies with the $\arctan(\omega nL/c)$ where ω is the optical frequency, n the index, c the speed of light and L a distance of propagation. Figure 1 shows examples of the time dependence of the pulse after different propagation distances when the phase varies with frequency as the $\arctan(\omega nL/c)$. Whereas the spectrum does not change with propagation, the temporal shape does as shown in Figure 1. Ultrafast processes that respond to the instantaneous field ought to be able to provide signatures of the temporal shape of the optical phase.

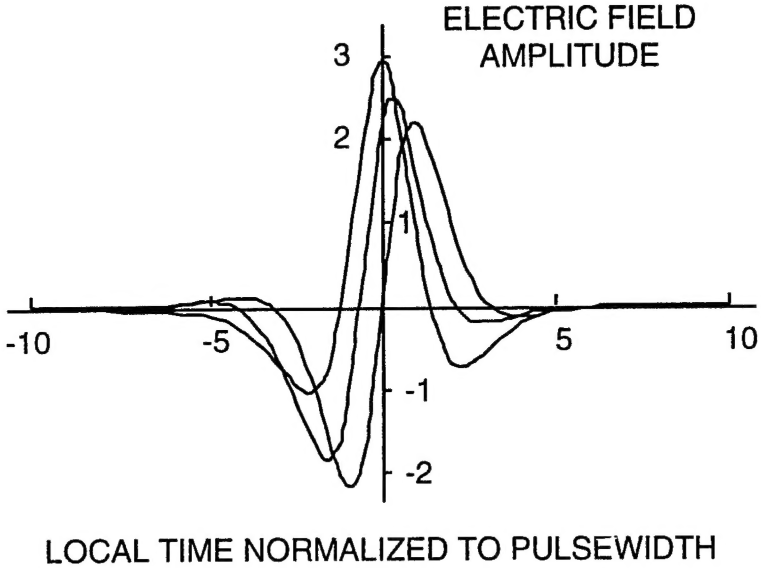

Figure 1: The change in the temporal dependence of the electric field amplitude as the phase evolves according to the $\arctan(\omega nL/c)$.

In a laser resonator an ultrashort pulse is shaped by the repeated action of both nonlinearity and dispersion. In practice, laser cavities are designed for careful control of the second order dispersion, and with as complete a cancellation as possible of the third and fourth order dispersions. This is accomplished with prism pairs introduced into the resonator, and/or properly designed dispersive mirrors. The roundtrip of the pulse envelope is controlled by the effective group delay (first order dispersion) and the phase shift from pulse to pulse by the phase delay. An indication of how different these can be is given by the following estimates: The propagation distance in sapphire that produces a π phase shift of the field oscillations under the envelope is 18.7 microns; for fused silica and air the distances are 29 microns and 9.8 cm respectively. So, for any system built to date, these numbers imply that the field oscillations shift dramatically with respect to the pulse envelope with each transit of the cavity.

Unless the resonator is designed and stabilized for a net roundtrip relative phase shift of zero (modulo 2π) successive pulses emitted from the laser will not have identical waveforms. Translation of the mirrors leaves the difference of the two delays unaffected. Control of the optical phase underneath the pulse envelope can only be controlled if the phase and group velocities are independently adjusted. Our group has recently

demonstrated that this is possible by translating of one of the prisms in the prism pair compensator. An alternative route to phase control has also been reported by L. Xu et al [4]. In order to verify the effectiveness of such control, the phase shift between successive pulses has to be measured in a pulse correlator. We did this using an interferometer with arms imbalanced by the distance between successive pulses. Relative phase changes as small as $1/20\pi$ between pulses were detectable. Figure 2 shows a fringe-resolved cross-correlation with a fringe peak shifted (by π) from zero delay by translating one of the intracavity prisms by 12 μm. It should be noted that the dispersion of air in one roundtrip of the long arm in the correlator is not negligible and its compensation via a thin glass slide was necessary. Thus far, we have not attempted to control the phase by feedback, and the "absolute" phase as referred to the pulse maximum was not determined.

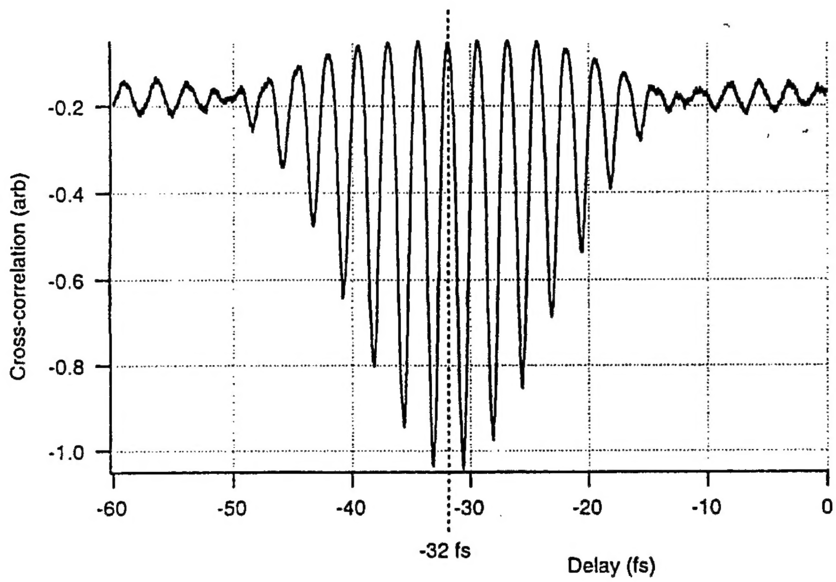

Figure 2: Fringe-resolved cross-correlation trace indicating π phase shift between two successive pulses from a 10 fs Ti:sapphire laser.

We propose to pursue the problem of ultrashort pulse generation and optical phase stabilization. One of the consequences of pulse waveform changes from pass to pass within the laser may be a limiting of the ultimate pulse duration. The amplitude stability of the laser may also be affected by the changing temporal relationship between envelope and phase. It is likely that stabilization of the optical phase will lead to greater amplitude stability from pulse to pulse. It is also possible that pulses even shorter than 6fs will be generated, if the phase stabilization scheme is successful.

We will investigate the generation of phase-controlled pulses in two different laser systems. The most direct approach, and the one with which our preliminary experiments have been carried out, is with Ti:sapphire. To make sure we can achieve the shortest pulses currently possible, we will use mirrors like those used in the record-setting

experiments of Prof. Keller's ETH Zurich group. The mirrors were, in fact, designed originally in collaboration with Prof. Haus [5]. Prof. Keller has agreed to make them available to us. We also propose to develop a Kerr-lens modelocked Cr:YAG for ultrashort pulses. The Cr:YAG system is of interest because it operates in the longer, 1.55 μm wavelength regime. To date the shortest pulses produced with this laser have been about 50 fs [6], but it has the gain bandwidth to permit pulses of 2 cycles (10fs) at these wavelengths. It also provides interesting opportunities for dispersion compensation since the dispersion of silica is anomalous in this regime. We have begun development of this laser and demonstrated self-starting with a quantum well saturable absorber.

With these new phase controlled sources we intend to investigate the effect of phase slippage on the generation process itself and on the amplitude fluctuations of the laser output. Ultimately the goal is to identify and study nonlinear ultrafast optical processes that demonstrate a sensitivity to the optical phase of the excitation or probing pulses. One example is a very thin back-biased tunneling junction excited by a beam incident upon it at an angle off the vertical. The field component normal to the junction will affect the tunneling in a way that depends on the details of the temporal field. The charge collected from each pulse should be an indication of the phase of the field. Another example is multiphoton ionization, which can depend strongly on the detailed temporal shape of the pulse.

This work will be performed in collaboration with Professors James Fujimoto and Hermann Haus.

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